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Article

Microfibrillated Cellulose Based Barrier Coatings for Abrasive Paper Products

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Abstract: Paper-based abrasive products are multilayer structures in which the first layer on the paper substrate is usually a latex barrier coating to prevent the migration of adhesive glue into the substrate. The high coat weight (10 g/m²) of latex barrier layers is a cause of environmental concerns. Hence, alternative materials that can provide the barrier function at lower coat weights are desired. In this work, microfibrillated cellulose (MFC) combined with poly(vinyl) alcohol (PVA) were explored as suitable alternatives to the current latex coatings. Barrier coating formulations containing PVA, MFC, and silica (SiO₂) were developed and applied to a paper substrate using a rod coating method on a pilot scale. Coating quality and barrier performance were characterized using scanning electron microscope images, air permeance, surface roughness, water contact angle, KIT test, and oil Cobb measurements. The barrier coatings were also studied for adhesion to the subsequent coating layer. An optimal barrier function was achieved with the developed coatings at a low coat weight of ca. 3 g/m². The adhesion of pure PVA and PVA-MFC barrier coatings to the subsequent coating layer was inadequate; however, silica addition was found to improve the adhesion.

Keywords: microfibrillated cellulose; polyvinyl alcohol; abrasive paper product; barrier coating; silica

1. Introduction

Abrasives such as sandpapers are usually multilayer structures, where synthetic polymer resins are binding and stabilizing abrasive particles on a backing sheet. Abrasive particles are inorganic materials, e.g., Al₂O₃ and SiC, of diverse sizes varying from 1 to 2000 µm according to the use. Several backing sheet materials such as papers, nonwovens, and plastics are used. The backing sheet material is often treated before use to improve its flexibility, durability, water resistance, and adhesion and barrier properties. Latex or other similar compounds are known to be particularly suitable for such treatments [1,2]. A combination of adhesive resin and an abrasive grit is applied to the treated backing followed by curing to yield an abrasive sheet material.

For paper-based abrasive structures, a barrier layer is applied on the substrate before the abrasive material coatings to prevent the migration of adhesive glue (synthetic polymer resins) used in the subsequent coating layers [3]. The barrier layer should also ensure proper adhesion of the subsequent coating layer [4]. These barrier coatings are currently made of oil-based latex with coat weights reaching almost 10 g/m². Recent environmental concerns related to oil-based products and rising demand for environmentally friendly products is driving the industry to search for sustainable alternatives to these barrier coatings, which can provide the desired barrier function at lower coat weights.

The development of barrier coatings for abrasive products has received surprisingly little attention from the research community. This is partly due to the existing commercial solutions of the abrasive products that have been used for a long time successfully without a need for further development.

However, there has been a lot of focus on developing sustainable biobased barrier coating solutions for paper packaging applications, as evident from several reviews on the topic [5–7]. Some of these developments from the packaging field can prove useful for the case here. For example, nanocellulose films and coatings with their known barrier performance against grease and oils [8–10] have the potential to replace the latex in the barrier layer for abrasive products. However, the high viscosity and yield stress of nanocellulose suspensions [11,12], even at low solid contents, create coating process challenges [13,14] and limit their use as the only material in the barrier layers for paper-based substrates. The high water content of the nanocellulose suspensions results in challenges related to shrinkage of the substrate, poor runnability, and high drying costs. However, other materials such as biodegradable polymer poly(vinyl) alcohol (PVA) with known barrier properties for oils [15], acceptable mechanical properties, and low cost can potentially be used in combination with nanocellulose to develop the barrier layer solutions. This could help in increasing the solids content of the coating formulation and minimize the challenges related to pure nanocellulose coating formulations. Furthermore, the resulting dry coating layer can be more flexible for further processing than nanocellulose coating alone.

PVA and nanocellulose combinations in films and composites have been explored by several researchers over the years [16–25]. PVA and nanocellulose coatings, on the other hand, are far less explored in general, and that too mainly for packaging [26,27] and printing applications [28]. Guezennec (2012) [27] proposed to use a PVA/cellulose nanofibrils (CNFs) layer as a barrier coating for developing a packaging board. A 10 g/m² coating layer comprising a 20:1 PVA/CNF ratio was deposited onto a 182 g/m² baseboard. The PVOH/CNF mixture was found to be better coatable than pure PVOH, which was due to the CNF helping during the drying by limiting blister defects and thus improving the runnability. According to the authors' knowledge, there have been no other studies exploring the use of PVA/microfibrillated cellulose (MFC) combinations in barrier coatings, especially for abrasive paper products.

The abrasive products use latex in the barrier coating, which not only provides a reasonable barrier but also ensures adequate adhesion to the subsequent coating layer. The adhesion is achieved both through mechanical and chemical interactions. The mechanical anchoring effect is obtained by a well-controlled migration of the glue into the substrate surface while the chemical binding of the latex to the glue improves the adhesion further. Therefore, the new barrier material replacing the latex should be multifunctional ensuring both suitable barrier and adhesion performance.

The objective of this work was to develop PVA and MFC based barrier coating layers for abrasive paper products. Three PVA based barrier coatings were produced, namely, pure PVA, PVA with the addition of MFC, and PVA with the addition of MFC and silicon dioxide where the latter was utilized as a filler to improve compatibility with the resin in the subsequent coating layer. The specific goal was to achieve the same barrier performance as the currently used latex barrier coating but with a lower coat weight while not compromising the adhesion to the subsequent coating layer. This would enable lean products and make abrasive material production more resource-efficient and sustainable in the future.

2. Materials and Methods

2.1. Materials

Mirka Ltd. (Finland) provided the paper-based substrate with a basis weight of 120 g/m². Borregaard AS (Sarpsborg, Norway) provided MFC, EXILVA F01-V, as 10 wt % water-based suspension/paste. PVA, POVAL® 6-98, was procured from Kuraray Europe Nordic Ab Oy (Vantaa, Finland) in the form of granules. Silicon dioxide (SiO₂), AERODISP® W1226, was procured as a 26 wt % water-based dispersion of AEROSIL® fumed silica from Evonik Resource Efficiency GmbH (Bitterfeld-Wolfen, Germany). Mirka also provided a latex coated barrier paper, which was used as an industry reference sample for comparing the performance of the new barrier coatings developed in this work.

2.2. Coating Formulations

Three coating formulations prepared in this work are listed in Table 1. The first formulation containing only PVA was prepared by cooking the PVA granules in the water at 85–90 °C for 2 h. For preparing the second formulation, the high viscosity MFC could not be dispersed efficiently in the cooked PVA solution. Therefore, a special protocol was developed to ensure proper dispersion of MFC in PVA. MFC was first dispersed in the total needed amount of water for the formulation, and PVA was cooked subsequently in the MFC dispersion at 85–90 °C for 2 h. This ensured an efficient mixing of MFC and PVA. For the third formulation, silica was added to a cold PVA-MFC mixture in a 70:30 weight ratio and mixed for 30 min. It is noted that the addition amounts of MFC and silica were first optimized on the laboratory scale. The MFC addition amount could not be higher because of the increasing viscosity of the formulation. The selected filler (silica) content was the maximum that provided good adhesion without compromising the barrier performance.

Table 1. Coating formulations.

Formulation	PVA	PVA-MFC	PVA-MFC-Silica
Weight ratio	100	90:10	(90:10)70:30
Solids content (%)	10	10	10

2.3. Rheology Measurements

Formulation rheology measurements with cone-plate geometry (plate diameter: 50 mm) were carried out using a Paar Physica Modular Compact Rheometer (MCR) 301 (Anton Paar GmbH, Graz, Austria). Shear flow tests were conducted in a shear rate range of 0.1–1000 s^{−1}.

2.4. Coating Application

The coating formulations were tested on a laboratory scale with an Erichsen laboratory coater before moving to the pilot-scale coating. Rod coating technology was utilized on VTT's surface treatment concept line (see Figure 1), and a 20 µm rod was used as a coating applicator. Coating speed was 10 m/min, and the coating was dried using infrared (IR) and air-drying. The coating width was 500 mm.

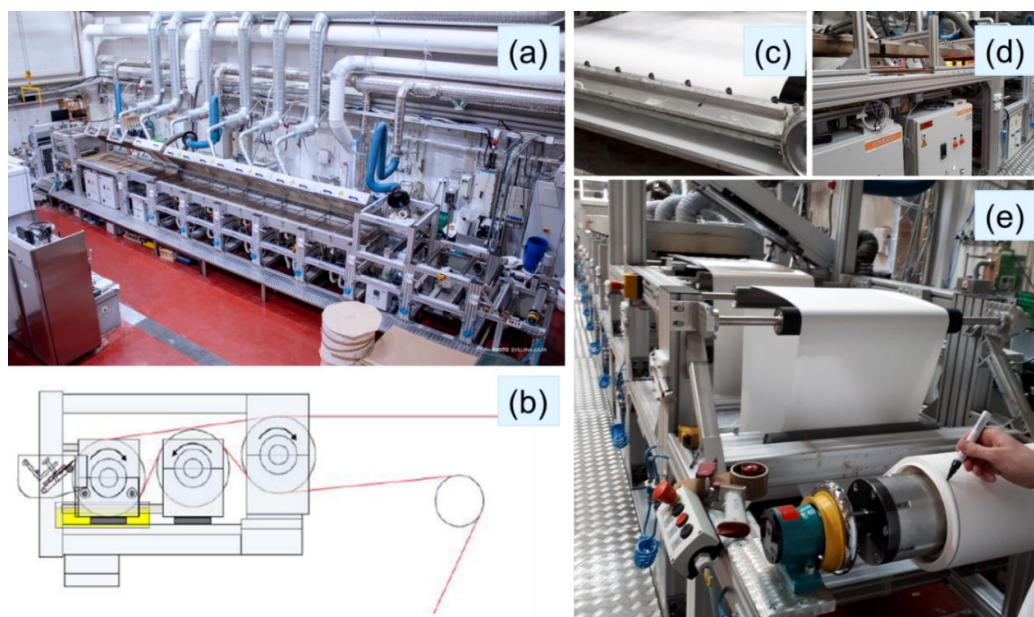


Figure 1. (a) VTT's surface treatment concept pilot line, (b) a schematic of the rod coating station, (c,d) coating application and drying, and (e) coated and dried paper.

2.5. Coating Characterization

2.5.1. Coat Weight, Air Permeance, and Surface Roughness Measurements

Grammage of the substrate and coated samples was determined following the TAPPI standard T 410 om-08 [29]. A precise determination of the low coat weights was difficult due to variation in the grammage of the substrate. Air permeance of the substrate and the coated samples was determined using an air permeability tester SE-166 (Lorentzen and Wettre, Kista, Sweden) with a measurement range of 0.003–100 $\mu\text{m}/(\text{Pa}\cdot\text{s})$ following the TAPPI standard T 547 om-18 [30]. The surface roughness of the substrate and coated samples was measured with an L&W PPS tester (Lorentzen and Wettre, Kista, Sweden) following the TAPPI standard T 555 om-15 [31]. For each property, five parallel determinations were made on different areas of each sample at 50% RH and 23 °C.

2.5.2. Water Contact Angle Measurements

The contact angle of water on the substrate and coated samples was determined by the sessile drop method employing a fully computer-controlled contact angle meter (KSV Instruments Ltd., Helsinki, Finland) equipped with a CAM 200 video camera. For each measurement, a water drop of volume 4.5 μL was placed on the surface of the sample, and images were recorded every second for 1 min. Up to five different positions on the sample surface were measured and the average contact angle determination is reported.

2.5.3. Oil Cobb Test with Special Monomer

Oil Cobb test was performed analogously to the water Cobb method (TAPPI T-441 standard method), where the testing liquid (water) was replaced by a monomer that was used as a component in the resin formulation of the subsequent coating layer.

2.5.4. KIT Test

The KIT test method, a standard method in the paper industry (TAPPI T559), was used to characterize the oil repellence of barrier-coated samples. KIT test was originally developed for testing the degree of oil and grease resistance of paper or board treated with fluorochemical sizing agents. The test method involves putting drops of a series of numbered reagents (KIT No. 1-12) on the sample surface. The highest numbered reagent (the most aggressive) that remains on the surface of the sample for 15 s without causing failure is reported as the “KIT number”. Testing of the barrier coated samples was done with the mixtures of castor oil and two solvents (n-heptane and toluene), where the higher solvent proportion provides a higher degree of aggressiveness. The average of five determinations for each barrier-coated sample is reported.

2.5.5. Oil Drop Test

An in-house developed oil drop test was used at VTT to determine the oil barrier of the coatings. The test was based on a visual estimation of a drop of red oil (rapeseed oil + red oil) coming through to the other side of the sample in 15 h.

2.5.6. Scanning Electron Microscopy (SEM)

Surface images of the samples were acquired with a Hitachi TM4000Plus scanning electron microscope (Hitachi, Japan) using a 100 times magnification.

2.5.7. Adhesion Tests

The barrier coating layers were tested for their adhesion to the subsequent coating layer. The test was performed by applying the next coating layer on the top of the barrier coating and then testing the

peel strength of the top coating. The peel strength was tested by rubbing the coated paper between the thumbs by rolling it back and forth. Thus, the adhesion test was of a qualitative nature.

3. Results and Discussion

3.1. Rheology and Coat Weights

The coating formulations were found to have different rheological behavior. The PVA formulation had the lowest viscosity, especially at low shear rates. The addition of MFC increased the viscosity, but the viscosity dropped again upon the addition of silica. The addition of MFC introduced a more pronounced shear thinning behavior to the formulation, as seen in Figure 2. The addition of MFC was also found to improve the coatability of the formulation probably by providing favorable rheological properties. The addition of silica to the PVA-MFC coating reduced the overall viscosity, but it did not affect the shear-thinning nature or coatability.

The coat weights obtained with the different formulations are listed in Table 2. Even though the formulations were at the same solids content, the higher coat weight for PVA formulation can potentially be explained by the difference in the rheological behavior of the formulations. The shear rates under the 20- μm rod at a coating speed of 10 m/min should be ca. 8000 s^{-1} . Based on the calculated power-law indices of the three formulations, the estimated viscosity at 8000 s^{-1} was lower for the PVA-MFC and PVA-MFC-silica formulations compared to the PVA formulation. The coat weight seems to follow the same trend as the shear-thinning index. The lower viscosity at high shear rates should also improve their coatability. It is noteworthy that the high viscosity of PVA formulation at high shear rates limits the minimum coat weight achievable with a 20- μm rod. However, the MFC helps achieve a lower coat weight with the same rod by bringing in its shear-thinning effect providing a lower viscosity at high shear rates. This is a clear advantage of using MFC in these formulations.

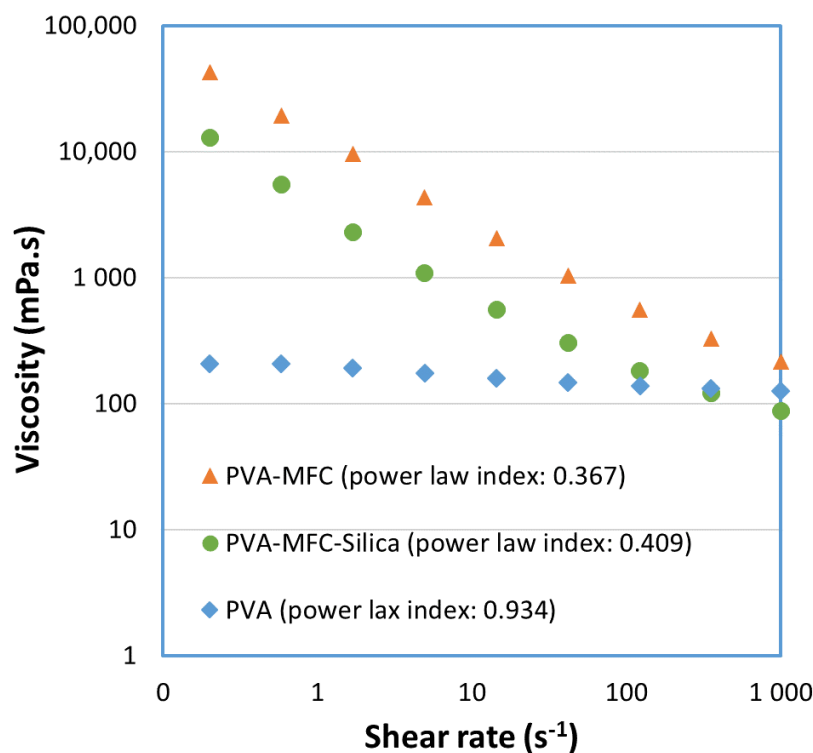


Figure 2. Viscosity against shear rate for the coating formulations.

Table 2. Coat weights and barrier properties obtained with different formulations.

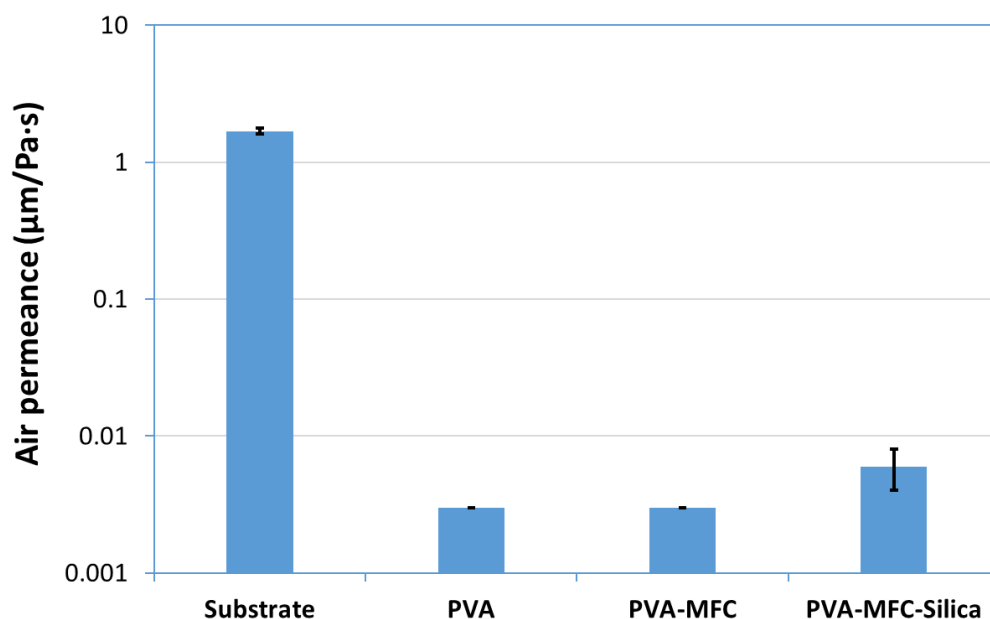
Sample	Reference	Substrate	PVA	PVA-MFC	PVA-MFC-Silica
Coat weight (g/m ²)	8–10	-	4	2.5	3
Air permeance (μm/Pa·s)	0.003 *	1.69 ± 0.09	0.003 *	0.003 *	0.006 ± 0.002
Surface roughness (μm)	5.5 ± 0.3	5 ± 0.3	6.7 ± 0.2	6.4 ± 0.4	7.1 ± 0.3
Oil Cobb (g/m ²)	3.8 ± 2	40.6 ± 1.4	2.1 ± 0.5	6.6 ± 1.3	8.5 ± 3.9
KIT number	9	0	11	9	6

* Detection limit of the instrument.

3.2. Barrier Performance

The air permeance of the substrate and coated samples are shown in Figure 3. Air permeance dropped significantly after coating implying a surface closure. The air permeance values for PVA and PVA-MFC coatings were below the detection limit (0.003 μm/Pa·s) of the instrument. The addition of silica somewhat reopened the coating structure, as indicated by the slightly increased air permeance.

All the coatings improved the oil barrier significantly compared to the substrate, as seen from Figure 4. The polar nature of the barrier coating components helps repel the non-polar oils. PVA coating demonstrated the best barrier performance, and the barrier deteriorated slightly upon the addition of MFC and silica. The water contact angles (see Table 3) of the barrier coatings show that similar hydrophilicity was achieved with all of them. Therefore, the barrier performance change seems to be happening due to the physical opening of the coating structure upon the additions of MFC and silica. Nevertheless, the oil Cobb values for all the coatings were found to satisfy the performance requirement. Furthermore, an oil drop test performed using a combination of rapeseed oil and red oil demonstrated good barrier performance for both PVA and PVA-MFC coatings because the oil did not come through to the other side of the coated samples in a testing time of 15 h. However, the PVA-MFC-silica coating did show some oil coming through to the other side probably caused by some pinholes. Nevertheless, all coatings showed excellent barrier at less than half the coat weight in the current latex-based solution, as seen from Table 2.

**Figure 3.** Air permeance of the substrate and coated samples.

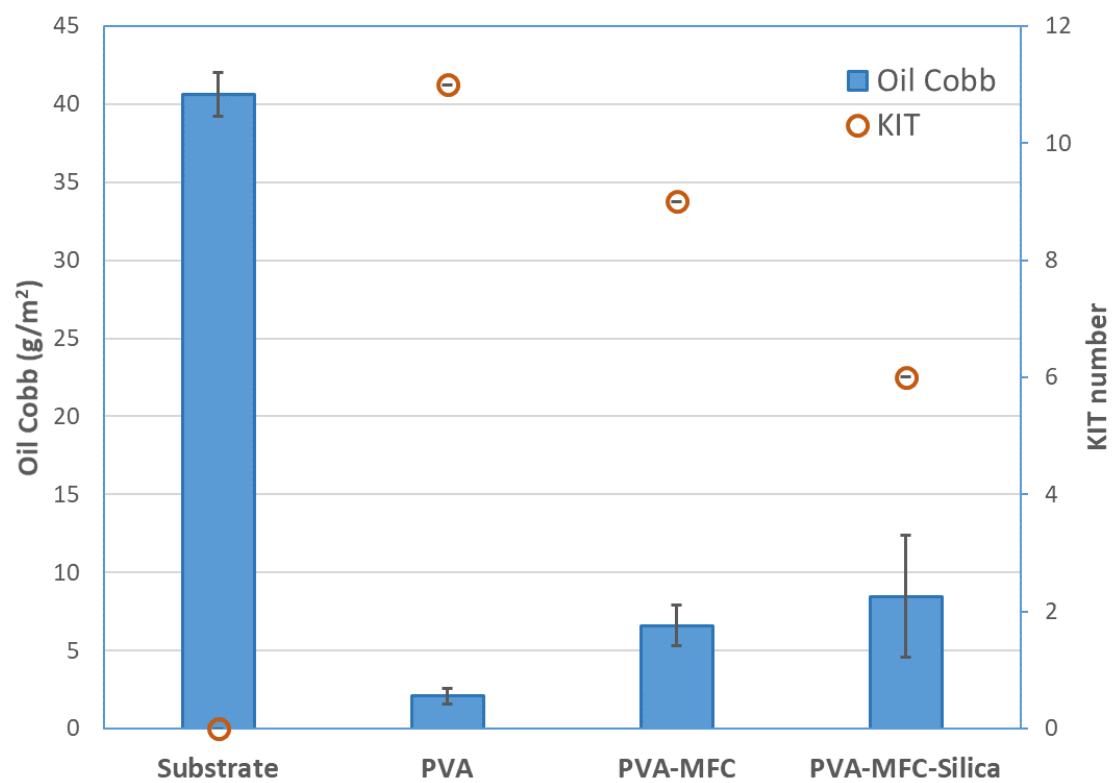


Figure 4. Oil Cobb and KIT test results for the substrate and coatings.

Figure 5 shows the surface SEM images of the substrate and coatings.

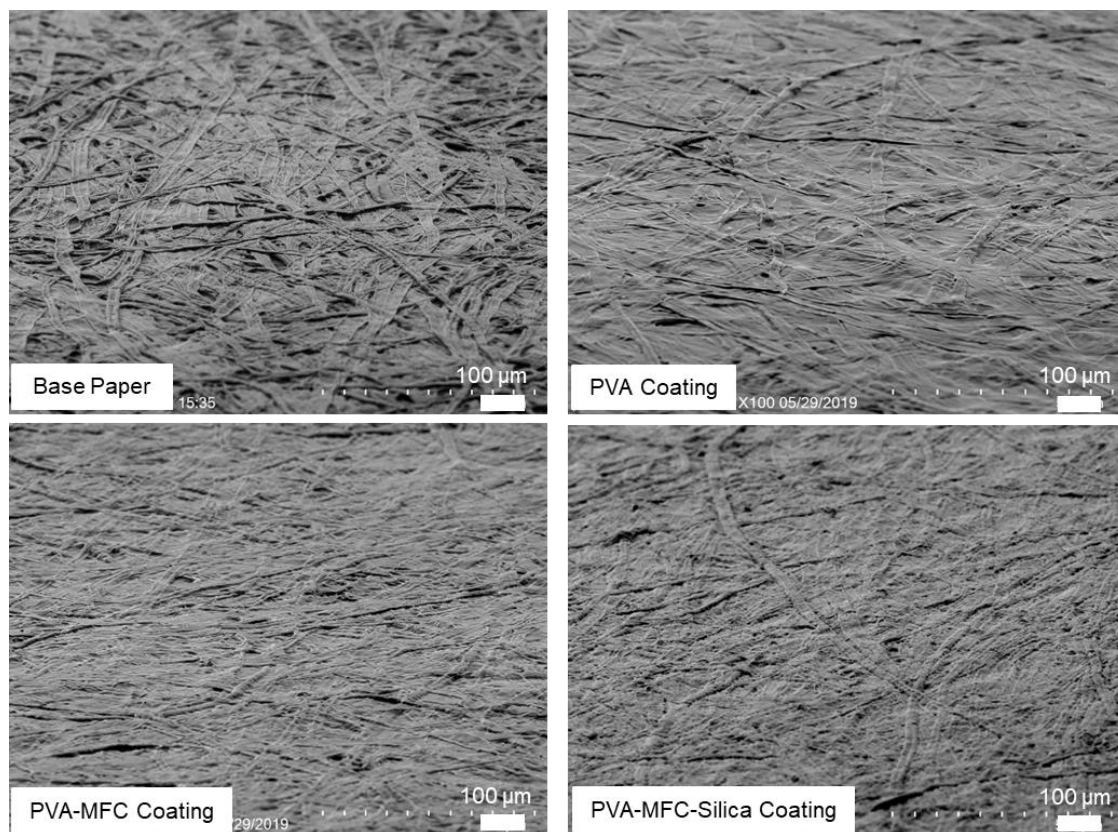


Figure 5. SEM images of the substrate and coating surfaces. Solid white bar denotes 100 µm.

As apparent from the SEM images and barrier performance, PVA coating clearly achieved a better surface closure. A slightly less closed surface coating was obtained with PVA-MFC. The addition of silica further opened the coating structure. A slightly open coating structure that meets the needed barrier requirements can be more beneficial when it comes to adhesion to the subsequent coating layer. The open coating structure can provide mechanical anchoring by allowing a controlled penetration of the monomer used in the subsequent coating layer.

3.3. Adhesion Performance

Table 3 lists the water contact angle of the substrate and coating layers. The coatings transformed the originally hydrophobic base paper into a hydrophilic surface. This was expected from the hydrophilic nature of the coating components involved. However, the water contact angle did not have a direct correlation with the adhesion to the subsequent coating layer. For example, all the coating layers had similar water contact angles but different adhesion performance. Therefore, the physical interaction of the coating components with the resin of the subsequent coating layer plays a more critical role. Furthermore, a relatively open coating surface in PVA-MFC compared to PVA coating seemed to improve adhesion through mechanical anchoring. Adhesion improved further in PVA-MFC-silica coating. This could be attributed to the opened coating structure and a potential chemical interaction of silica itself with the monomer. Therefore, silica seemed to act as an adhesion promoter, as also seen from Figure 6. There was a clear failure of the next coating structure applied to PVA and PVA-MFC barrier coatings. Only the PVA-MFC-silica coating provided significantly improved adhesion.

Table 3. Water contact angles of the coatings and adhesion performance.

Sample	Water Contact Angle at 0.5 s (°)	Adhesion to the Next Layer (Qualitative Estimate)
Substrate	112	-
PVA	58	Poor/No adhesion
PVA-MFC	46	Poor
PVA-MFC-Silica	55	Good

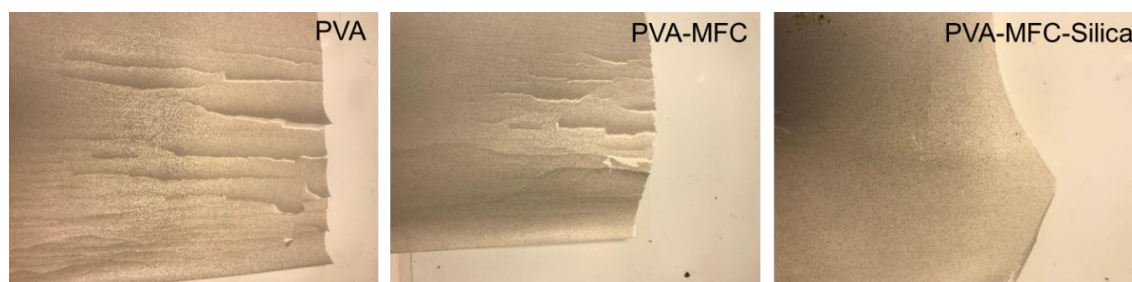


Figure 6. Coated samples after performing the adhesion test.

Surface roughness is also an important aspect of mechanical anchoring of the next layer. Surface roughness, shown in Figure 7, also seemed to correlate with the coating coverage and adhesion performance.

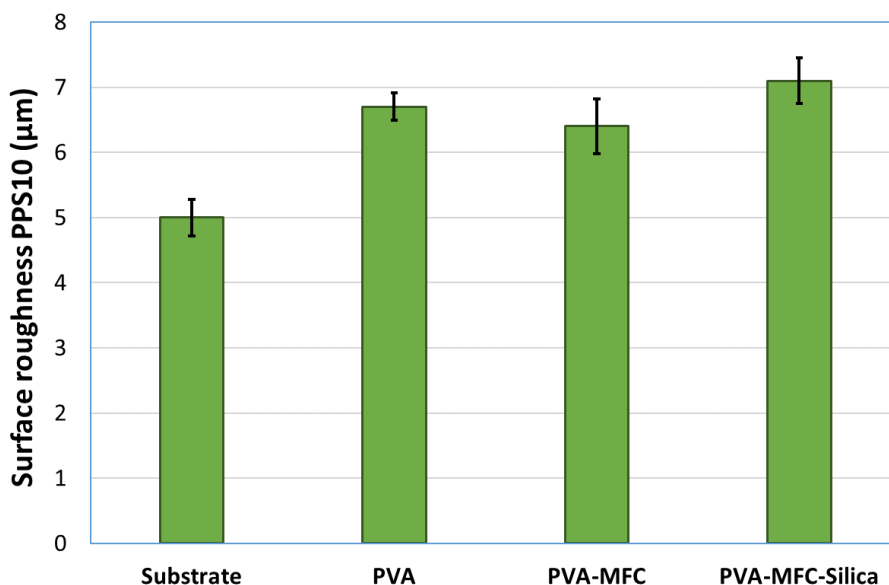


Figure 7. Surface roughness of the substrate and coated samples.

The increase in surface roughness was the highest for silica-containing coating, which can prove helpful in mechanical anchoring of the subsequent coating layer. The swelling induced during coating probably caused increased surface roughness with all coatings compared to the base paper. However, some curling of paper due to barrier shrinkage was observed, and this needs to be compensated in future coating trials. A common solution is to add some water to the other side of the paper to prevent the curling.

4. Conclusions

New barrier coatings containing PVA, MFC, and silica for abrasive products were developed and demonstrated at the pilot scale. MFC addition was found to improve the coatability and reduce the final coat weight of the formulations by providing favorable rheology. Excellent barrier performance was achieved at a much lower coat weight than the currently existing latex-based solutions. The polar nature of the coating components played a key role in achieving the barrier performance against oil-based synthetic resins. However, a fully closed coating structure with PVA prevented essential adhesion of the subsequent coating layer. A slightly open coating structure, achieved with the addition of both MFC and silica, reduced the barrier performance but increased the adhesion to the subsequent coating layer by providing a mechanical anchoring effect. These new barrier solutions could pave the way for the development of abrasive products with lower environmental impacts than with oil-based solutions.

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